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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Composition of cirrus-forming aerosols at the tropical tropopause" *by* K. D. Froyd et al.

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We thank the anonymous referee for their careful review and insightful comments. Below are referee comments and author responses.

Specific Comments 1) Title: Since these measurements are related to subvisible cirrus and its formation and chemistry and may be very different from other (more studied) types of cirrus, it would be useful to include the term "subvisible cirrus" somewhere in the title.

Response: The title was changed to, "Aerosols that form subvisible cirrus at the tropical tropopause"

2) p. 20349, 1st paragraph: Since heterogeneous and homogeneous freezing are also

dependent on temperature as well as supersaturation, please include typical temperatures at which subvisible cirrus forms in this paragraph.

Response: Temperature ranges for the RH observations were added.

3) p. 20350, line 25: crystal “size” should be further defined: I assume it is an optical diameter (not radius), but is it maximum length, mean diameter, or something else?

Response: “Size” was clarified as "diameter (maximum dimension)".

4) p. 20351, lines 10-15: Some more details would be useful to better understand the authors’ reasoning here. The discussion is about artifacts in convection and anvil cirrus. The main measurements presented in the paper, however, refer to subvisible cirrus. The authors seem to imply earlier that the crystals in such clouds are small enough to be efficiently sampled (presumably) without contact with inlet surfaces. So, it would be useful to note initially in this discussion that other types of cirrus were also sampled (as inferred but not really detailed with respect to Fig. 2.) Also, it is stated that secondary particle generation was “significantly reduced” by plating the inlet with gold. It would seem difficult to prove this without sampling with and without the gold plating in clouds with similar crystal sizes and types (and airspeeds). Was this done, and if so, is it presented elsewhere? If not, those data should be presented here to support the authors’ statements.

Response: The text was changed and a reference added to clarify that sampling artifacts were observed by PALMS within non-SVC cloud during current and previous studies. “Not only do ice crystals shatter and alter the measured cloud particle size distribution (Jensen et al., 2009a), but crystals with enough mass ablate pieces of inlet material that can then be mistaken for cloud residuals (Murphy et al., 2004). During previous PALMS flight studies within anvil cirrus (Cziczo et al., 2004a), stainless steel particles were efficiently generated from crystal impaction on the CVI inlet. These micron-sized contamination particles can be identified with composition-resolving instruments such as PALMS or electron microscope analysis provided the technique is

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sensitive to the contaminant."

Also, the text was clarified regarding the effect of gold coating: "For the current study, CVI inlet surfaces were plated with gold. In laboratory studies (Murphy et al., 2004), secondary particle production from ice impaction onto gold was significantly lower than onto stainless steel, presumably due to the more ductile nature of gold. Consequently, contamination data rates within anvil cirrus during CR-AVE were reduced compared to previous deployments."

5) Fig. 1 gives an example of chemical spectra for three different types of contamination particles. Fig 1c shows trace metals (Cu and Sn) that are given as an indication of contamination with mineral dust, yet these are also present in the earth's crust; if these are assumed to be from the inlet, what is their source within a gold-plated inlet?

Response: Contamination signatures are identified not only by their elemental composition but also the relative intensity. In Fig 1c, Cu+ and Sn+ signatures are 10-100 times more intense than observed in mineral dust particles. The source of these metals is either ablation of not only the gold coating but also the stainless steel beneath, or contamination within the gold coating itself introduced during the electroplating process. The presence of Fe+ and Cr+ in this spectrum suggests that the first possibility may be most applicable here, although Fe+ and Cr+ cannot be unambiguously identified as contamination signatures since these can be present in mineral dust at similar intensities.

6) Fig. 2 gives the percentage of total particles influenced by contamination as a function of ice water content, which is interesting. Since the authors discuss several different potential types and sources of contamination, to aid in understanding the results, please include the relative percentages of these different contamination types detected.

Response: Contamination fractions for each category of contamination particle were added to section 2.

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7) p. 20353, line 23: Particles with ammonium to sulfate ratios of 3:2 or 2:1 are given as the most common, but Table 1 says the ratios for all samples were greater than or equal to 2. Do you mean less than or equal to 2?

Response: In the paragraph starting, “In a recent study (Froyd et al., 2009),...” the 3:2 or 2:1 ratio refers to the typical background particles throughout the TTL. In Table 1, we report the average NH4+:SO42- ratios for unfrozen aerosols during the specific SVC events detailed in the current study. The $\geq$2 value is correct.

8) p. 20354, line 20-21: I don’t understand why it is stated that mineral dust was not enhanced in cirrus residues unless I am misinterpreting Table 1, dust is actually a higher percentage of cirrus residue samples than of unfrozen aerosol samples. Is it because the number of dust particles are insufficient to be statistically significant? If so, the statistics should be presented.

Response: Compared to large mineral dust enhancements ($\sim$30-50x) in anvil cirrus shown in Fig 4 and detailed in the Cziczo et al. 2004b reference, dust enhancements in SVC residue are probably small enough to be negligible (2 in 98 vs. 1 in 358). The limited sample population is mentioned in the text and apparent from Table 1. The text was changed to read: “Lower tropospheric aerosols such as mineral dust were not significantly enhanced in subvisible cirrus residues.”

9) p. 20354, line 25: Suggest inserting “heterogeneous” IN, if that is what you mean here.

Response: “heterogeneous” inserted.

10) p. 20355, line 15-17: Wouldn’t the Murphy et al. 2006b results be from a different field project? Perhaps they are assumed to be relevant since sampling was conducted in the same region, but this should be clarified.

Response: The Murphy et al. 2006b study establishes that mercury and halogens accumulate onto aerosols at the cold temperatures of the tropopause region. That

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study was from a different field project, but the accumulation mechanism is described as dependent primarily on temperature and should therefore be relevant to other (and colder) tropopause locations.

11) p. 20356, line 19 (discussion of cirrus concentrations): Fig. 5c is helpful in showing the size of particles detected by PALMS, which is only the far end of the accumulation mode. Can the authors give an estimate, based on the sampling rate and efficiencies, of what fraction of the subvisible crystal number concentration was actually sampled by PALMS and so is actually represented by these results?

Response: This calculation requires several estimations and therefore has large uncertainties. However, we have determined a rough estimate and have added this statement to section 2: "It is difficult to calculate the fraction of SVC crystals detected by PALMS as residuals. An accurate determination would depend on several high-order factors including the sub-isokinetic inlet aspiration efficiency, ice crystal stopping distances and heating rates, PALMS size-dependent detection efficiency, and the unknown size distribution of SVC residues. For a rough indication of SVC detection, we convert the detection rate for SVC residuals of 0.062 Hz into an ambient concentration. By then comparing to ambient crystal concentrations, we estimate that PALMS is detecting residuals for $\sim$2% of 5-25 um SVC crystals. This number is within the PALMS detection efficiency range for particles >250 nm (<1% to $\sim$10%) and provides some confidence that a substantial fraction of the residuals are within the PALMS size range. It is expected that residuals detected using the current method are representative of the residual population and not a special subset."

12) p. 20357, lines 9-15: Is there any evidence, through in-situ aerosol shape/phase measurements or environmental SEM, that the ambient particles actually were dry in this environment? That would be something interesting to measure in the future.

Response: There was no indication from PALMS optical scattering signals relative to measured aerodynamic diameters that frozen aerosols had a non-spherical shape, al-

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though this effect would be subtle and would require a large sample population. As a historical note, Bigg and coworkers (Bigg, 1986; Gras, 1978) reported sporadic evidence of dry aerosols in the mid-latitude lower stratosphere from jet impaction patterns recorded aboard balloon flights. The following text was added to section 4: "Impactor samples from the subtropical stratosphere have occasionally yielded electron microscope images indicative of dry ammonium sulfate."

13) Fig. 3 caption: How was IWC calculated from both 2D-S channels? Were they just averaged?

Response: The channels were averaged. This was clarified in the caption.

14) Fig. 4b: Please include the number of anvil cirrus residuals represented by the pie charts, as these don't seem to be given in Table 1.

Response: Residual totals were added to Figure 4b.

Minor Technical Corrections 15) p. 20357, line 22: Delete extra "and".

Response: Done.

References Bigg, E. K.: Ammonium Compounds in Stratospheric Aerosols, Tellus, 38B, 62-66, 1986. Gras, J. L.: Change in Nature of Stratospheric Aerosol Collected at 34-Degrees-S, Nature, 271, 231-232, 1978.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 20347, 2009.